Supporting information

Spin-reorientation-induced magnetodielectric effects in two layered perovskite magnets

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Fig. S1 Photos of crystals of PEA-Mn and PEA-Cu view along the *c*-axis.



Fig. S2 The simulated, experimental powder and single crystal (along the *c*-axis) XRD patterns of PEA-Mn and PEA-Cu.



Fig. S3 The hydrogen bonds act as bridges between the organic and inorganic layers in the crystal structures of PEA-Mn and PEA-Cu. The phenethyl groups of PEA were omitted for clarity. The hydrogen bonds are represented by dashed lines.



Fig. S4 Zero-field-cooled magnetization (ZFCM) and field-cooled magnetization (FCM) under 20 Oe for a crystal sample of PEA-Cu along the *c*-axis.



Fig. S5 Magnetizations of PEA-Mn (a) and PEA-Cu (b) at different temperatures measured by applying magnetic field on a single crystal along the *c*-axis.